LANSCE
Los Alamos Neutron Science Center

Research Proposal for the use of Neutron Science Facilities

☐ Fast Access ☐ Joint CINT Proposal

Proposal Number: 20111529
Submission Number: S1539
Date Received: 03/09/11

Program Advisory Subcommittee: Basic Nuclear/Particle Physics
Focus Area:

Flight Path/Instrument: 1FP14 / DANCE
Dates Desired: 10/20/2011 - 12/21/2011
Estimated Beam Time (days): 16
Impossible Dates:
Days Recommended: 0

TITLE
Neutron capture measurements on 173Lu at DANCE

☒ Continuation of Proposal #: 2009-S1222
☒ Ph.D Thesis for: THEROINE Camille

Principal Investigator: Roig, Olivier
Institution: Commissariat a l'Energie Atomique-Bruyeres-le-Chatel
Citizenship: France
Phone: 01133169264140 **FAX:**
Email: olivier.roig@cea.fr
Local Contact: Ullmann, John L.

Co-Proposers	Institution	Citizenship	E-mail Address
Bail, Adeline	Commissariat a l'Energie Atomiq	France	adeline.bail@cea.fr
Bond, Evelyn M	Los Alamos National Laboratory	United States of Am	bond@lanl.gov
Bredeweg, Todd A	Los Alamos National Laboratory	United States of Am	toddb@lanl.gov
Couture, Aaron	Los Alamos National Laboratory	United States of Am	acouture@lanl.gov
Daugas, Jean-Michel	Commissariat a l'Energie Atomiq	France	jean-michel.daugas@cea.fr
Haight, Robert C	Los Alamos National Laboratory	United States of Am	haight_robert_c@lanl.gov
Jandel, Marian	Los Alamos National Laboratory	Slovak Republic	mjandel@lanl.gov
Meot, Vincent	Commissariat a l'Energie Atomiq	France	vincent.meot@cea.fr

RESEARCH AREA

☐ Biological and Life Science
☐ Chemistry
☐ National Security
☐ Earth Sciences
☐ Engineering
☐ Environmental Sciences
☒ Nuc. Physics/chemistry
☐ Astrophysics
☐ Few Body Physics
☒ Fund. Physics
☐ Elec. Device Testing
☐ Dosimetry/Med/Bio
☐ Earth/Space Sciences
☐ Materials Properties/Test
☐ Other:

☐ Mat'l Science (incl Cond Matter)
☐ Medical Applications
☐ Nuclear Physics
☐ Polymers
☐ Physics (Excl Condensed Matter)
☐ Instrument Development
☐ Neutron Physics
☐ Fission
☒ Reactions
☐ Spectroscopy
☐ Nuc. Accel. Reactor Eng.
☐ Def. Science/Weapons Physics
☐ Radiography
☐ Threat Reduction/Homeland Sec.
☐ Other:

FUNDING AGENCY

☐ DOE/BES
☐ DOE/OBER
☒ DOE/NNSA
☐ DOE/NE
☐ DOE/SC
☐ DOE/Other

☐ DOD
☐ NSF
☐ Industry
☐ NASA
☐ NIH
☒ Foreign: CEA/DAM
☐ Other US Gov't:
☐ Other:

PUBLICATIONS

Publications:

1FP14 - DANCE
____
- O. Roig for the DANCE collaboration, "Neutron capture reactions on Lu isotopes at DANCE", in Procceding of the 12th International Conference on Nuclear Reaction Mechanisms, Villa Monastero, Varenna, Italy, 15 - 19 Jun 2009, CERN-Proceedings-2010-001, pp.217-222 (2010)
- O. Roig and A.J. Couture, "Neutron capture reactions on Lu isotopes at DANCE", in Procceding of the 10th Symposium on Nuclei in the Cosmos, Mackinac Island, Michigan, USA, 27 July – 1 August, 2008, PoS(NIC X)077 (2008)

IPF
______
- W.A. Taylor et al, "Production of a 173Lu target for neutron capture cross section measurements", J. Radioanal. Nucl. Chem. 282, pp. 391-394 (1009)

Abstract: S1539_Proposal_LAN.pdf

By electronic submission, the Principal Investigator certifies that this information is correct to the best of their knowledge.

Safety and Feasibility Review *(to be completed by LANSCE Instrument Scientist/Responsible)*

☐ **No further safety review required** ☐ **To be reviewed by Experiment Safety Committee**

☐ **Approved by Experiment Safety Committee, Date:**

Recommended # of days:	**Change PAC Subcommittee and/or Focus Area to:**	**Change Instrument to:**

Comments for PAC to consider:

Instrument scientist signature: **Date:**

Neutron Capture Measurements on ^{173}Lu at DANCE

D.J. Vieira, T.A. Bredeweg, E.M. Bond, M. Jandel,
R.S. Rundberg, J.B. Wilhelmy (C-NR)
F.M. Nortier, W.A. Taylor (C-IIAC)
J. L. Ullmann, A. Couture, J.M. O'Donnell, R.C. Haight (LANSCE-NS)
J.M. Wouters[†] (IST-AUBAD)
Los Alamos National Laboratory

O. Roig, A. Bail, J.-M. Daugas, V. Méot, P. Morel, C. Theroine
CEA Bruyères-le-Châtel, France

I. Introduction

Herein we propose to continue the measurement of the neutron capture on a radioactive Lu target composed of ^{173}Lu ($t_{1/2}$=1.37 yr), using the DANCE array on flight path 14. This work benefits from measurements that made on isotope-enriched stable targets of ^{175}Lu and ^{176}Lu that ran in 2007 and being published. This proposal is also following two previous test-experiments which took place in 2008 and 2009. From the experience of these two test-experiments, we would like to request 2 day of setup and testing time to install the new radioactive target holder in DANCE with a new designed lead liner, 8 days of beam time to acquire production data with the radioactive target, 6 days for background (blank and natural Yb) and Au calibration runs, for a total beam time request of 16 days.

II. Motivation

Selected radiochemistry detectors have been used in the NTS test program to obtain neutron fluence and performance information. High energy neutrons, above the Q-value for (n,2n) reactions produce neutron deficient species (A-1 and A-2) from the radiochemistry detector isotope (A, if mono-isotopic). Low-energy or "late-time" neutrons that induce neutron capture reactions will reset to some extent the isotope ratio of (A-2)/(A-1) reaction products that are measured. Thus to correctly interpret these radiochemistry detector isotopic ratio results, we must know the neutron capture cross sections. Herein we propose to continue the investigation of one radchem detector --- Lu.

Although the most important measurements are the destruction cross section measurements on the A-2 or A-1 (i.e. radioactive) species, we have started with the Lu natural isotopes in last years experiment. This measurement proved to be very successful and so in 2008 we proposed to begin measurements on the radioactive isotopes - ^{173}Lu and ^{174}Lu. Two experiments, in 2008 and 2009, were necessary to investigate background and measurements problems that are associated with radioactive target and to optimize the target and experimental configuration. The first ^{173}Lu target used in the first measurement attempt in 2008 was contaminated by ^{88}Y much more than manageable (Figure 1). A more purified ^{173}Lu target including a chemical separation of stable Yb β-daughters and others rare earth isotopes was used in 2009. For this proposal, we will use this second target. However, a rare earth chemical separation that will remove the stable Yb isotopes will be achieved just before the run.

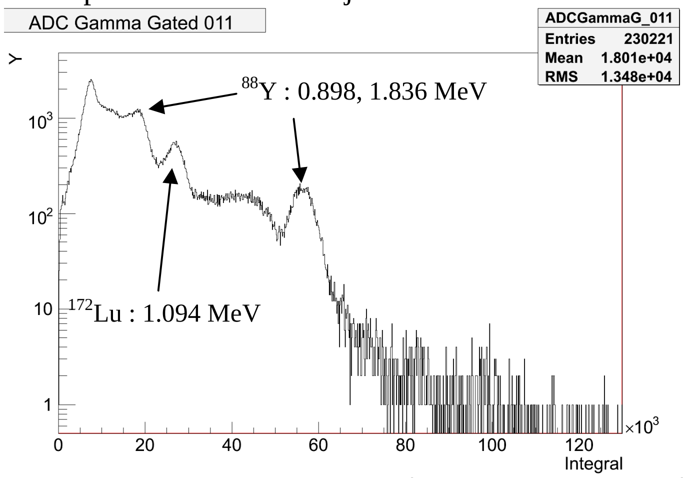


Figure 1: <u>Run 2008</u>, gamma energy spectrum for a single BaF_2 crystal of DANCE

Related to the experimental configuration, DANCE performance was affected by the activity of the target but problems of DANCE working conditions were minimized by the use of a tungsten shield around the target. A tungsten liner was preferred to a lead liner, initially proposed, because of its good gamma attenuation property and its capacity to be machinable as we wanted.

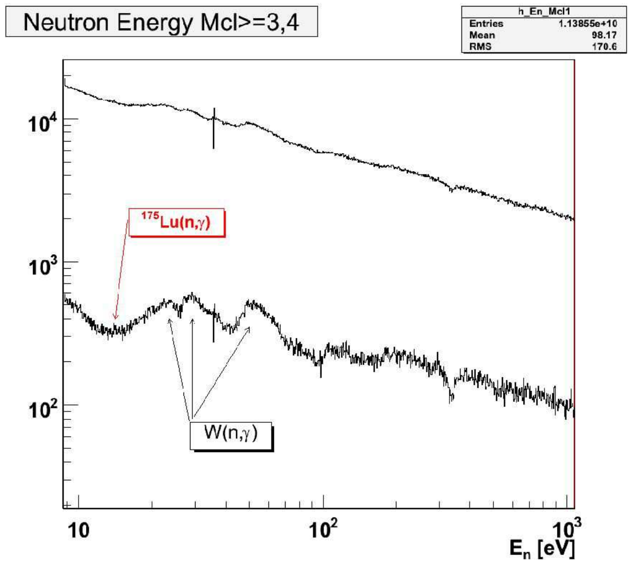


Figure 2: <u>Run 2009</u>, Counts versus neutron energy in eV range. Neutron resonances due to the presence of the W liner are marked.

Nevertheless, the interaction between scattered neutrons and a liner of the ^{173}Lu target was underestimated. We have observed, from the second experiment analysis, tungsten neutron resonances (Figure 2), largely visible as a background in the eV range. Resonances are broadened as the scattered neutron energies were determined through a capture event along the 30 cm long liner.

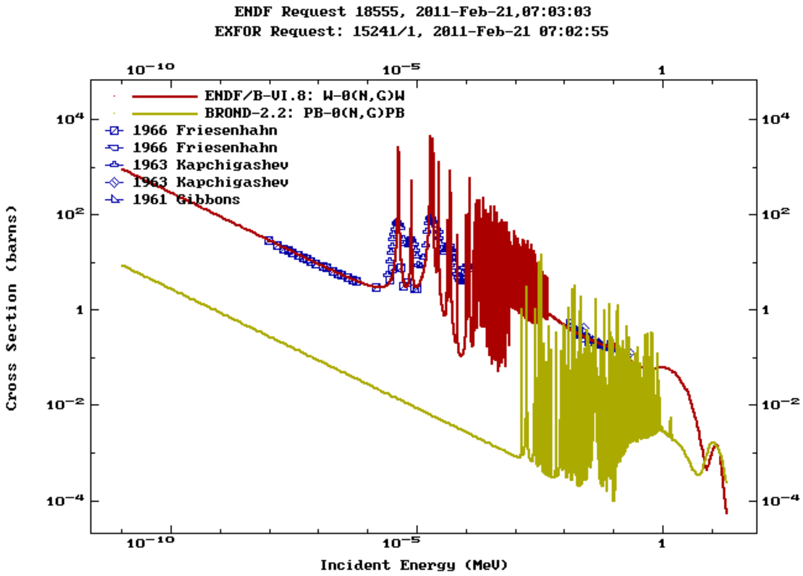


Figure 3: Neutron radiative capture cross sections for Pb and W using the ENDF/EXFOR data.

As, combining a highly radioactive and very small (60 µg) target, data above the keV region seems to be too challenging to get, we can use a Pb liner having a smaller (100 times less) cross section in eV region. This beam time request will enable us to investigate the neutron resonance region. Moreover, since it is likely that some amount of stable isotopes will be present in the radioactive targets, we will use last years data to subtract out the stable isotope contribution. In this way, we will be able to obtain the first measurements of neutron capture on ^{173}Lu.

For additional information on past DANCE proposals related to Lu neutron capture, please refer to:

(1) #2006-2542, entitled “Neutron Capture Measurements on ^{169}Tm, ^{175}Lu, and ^{75}As at DANCE”;
(2) #2007-S1201, entitled “Neutron Capture Measurements on ^{75}As, ^{175}Lu, and ^{176}Lu, at DANCE.”
(3) #2008-1540, entitled “Neutron Capture Measurements on ^{173}Lu, ^{174m}Lu and ^{174}Lu, at DANCE.”
(4) #2009-S1222, entitled “Neutron Capture Measurements on ^{173}Lu, ^{174m}Lu and ^{174}Lu, at DANCE.”

III. Experiment Details and Beam Time Request

a. ^{173,174}Lu target preparation

To prepare the ^{173,174}Lu radioactive target, we use proton-induced reactions on a Hf target irradiated at the isotope production facility (IPF) at LANSCE. As the figure below shows, the ^{nat}Hf target consists of several isotopes, on which (p,xn) and (p,pxn) reactions will lead to neutron-deficient isotopes of Ta and Hf. Among these isotopes, two mass chains can beta-decay to ^{173}Lu and ^{175}Lu, while ^{174}Lu is shielded by stable ^{174}Hf. However, with 70-90 MeV protons used in the irradiation, we also expect some direct production of Lu isotopes by (p,2pxn) reactions. Via this path there will also be some ^{174}Lu and ^{176}Lu produced in addition to the other isotopes mentioned above. Thus upon chemical remove of the Lu fraction from the Hf target using the C-Division hot cells, we will obtain a mixed isotopic target containing $^{173-176}$Lu.

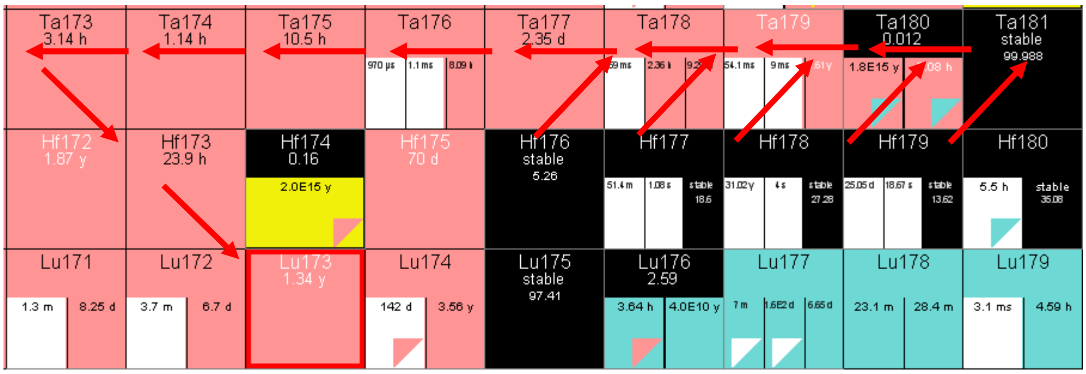


Figure 4: Proton-induced reactions on a Hf target leading to the production of a Lu radioactive target following the red arrows.Black squares are stable isotopes. Red squares are for neutron-deficient unstable isotopes. Green squares are for neutron-rich unstable isotopes. Natural abundance, half-lives are written inside the squares.

A ~50 g Hf target was irradiated with 70-90 MeV protons at 200 µA average current for one week at Isotope Production Facility (IPF) near the end of 2008. Based on ALICE calculations and the previous target production, we expect to have produced enough ^{173}Lu to make several radioactive targets on the order of 100 µg/cm^2. We are presently in the process of assaying the irradiated target after a five month cooling time to determine it isotopic composition. The Hf-Lu radiochemical separation has been done before (both at LANL and BIII), but we needed some time to reestablish and refine this separation which was done in a hot cell. This work was published in [W. A. TAYLOR *et al.*, J. Rad. Nucl. Chem., **282**, pp. 391-394 (2009)]. For this proposal, we will only need to process the last rare earth chemical separation that will remove the stable Yb isotopes, β-daughters of ^{173}Lu. Then, the target will be electroplated onto two thin titanium foils that are then mount and glued together on a DANCE target frame.
For the moment, we assume a ^{173}Lu radioactive target including a negligible part of ^{174}Lu (4‰) and stable Lu isotopes.
In particular, the radioactive species in the target are expected to be:

- ^{173}Lu ($t_{½}$ = 1.37 years, $J^{\pi} = 7/2^{-}$)

- ^{174g}Lu ($t_{½}$ = 3.31 years, $J^{\pi} = (1^{-})$)

as well as some some stable ^{175}Lu and ^{176}Lu.

b. DANCE measurements

From the test-experiments and the radioprotection limit to prepare and to handle the target, 100 mRem/h at 30 cm, we are aiming for a target that will have an areal thickness of 200 µg/cm^2 deposited over a diameter of 6.35 mm that is sandwiched between two 2.5 µm (1.2 mg/cm^2) Ti backing foils.
Using the isotopic/isomeric assumptions given above, we expect the target to contain the following composition and activity levels at the time of the experiment (see Table 1). Additional tables are given for individual γ-rays and their intensities after 5 mm, 1 cm lead attenuators surrounding the target for ^{173}Lu (Table 2).

Table 1: *Expected composition and activity levels of the radioactive ^{173,174}Lu target at the experiment time (June 2011) for a* ***200 µg/cm^2 target****.*

Target	***^{173}Lu***	***^{174g}Lu***	***Total***
Mass (mg)	0.064	0.00026	
Atoms number (atoms)	2.229×10^{17}	9.002×10^{14}	
Activity (Bq)	3.664×10^{9}	5.555×10^{6}	
Gamma dose rate at 30 cm (mRem/h)	107.9	0.1074	**108**

Table 2: *The estimated gamma-activity for specific lines in ^{173}Lu given the quantities list in Table 1. Bold letters are used to highlight the most intense γ-lines for a* ***200 µg/cm^2 target****.*

γ-rays from ^{173}Lu	***Intensity***	***After 5 mm Pb***	***After 10 mm Pb***
62.17 keV	0.17%	3.27×10^{-5}	0
78.63 keV	11.90%	263	1.58×10^{-4}
100.72 keV	5.24%	7.33×10^{-6}	0
171.39 keV	2.90%	2.86×10^{4}	7.68
179.37 keV	1.38%	3.22×10^{4}	20.5
223.16 keV	0.14%	6.44×10^{4}	80.7
233.61 keV	0.55%	3.91×10^{5}	7.6×10^{3}
272.11 keV	**21.20%**	$\mathbf{4.68\times10^{7}}$	$\mathbf{2.82\times10^{6}}$
285.36 keV	**0.61%**	$\mathbf{1.77\times10^{6}}$	1.39×10^{5}

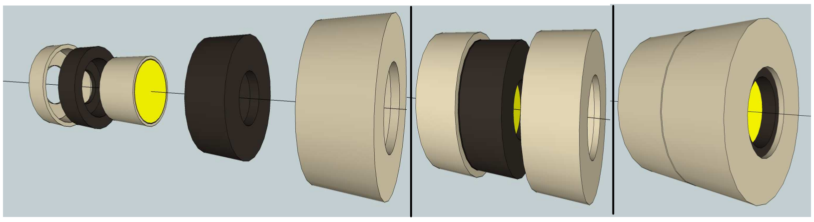
γ-rays from ^{173}Lu	*Intensity*	*After 5 mm Pb*	*After 10 mm Pb*
334.32 keV	0.11%	6.41×10^{5}	1.02×10^{5}
350.77 keV	**0.30%**	**2.06×10^{6}**	3.85×10^{5}
456.79 keV	0.14%	**1.79×10^{6}**	6.24×10^{5}
557.5 keV	**0.52%**	**8.72×10^{6}**	**3.99×10^{6}**
636.11 keV	**1.45%**	**2.76×10^{7}**	**1.43×10^{7}**
Gamma activity (γ/s)		**8.99×10^{7}**	**2.24×10^{7}**
DOSE RATE at 30 cm (mRem/h)		**8.29**	**3.21**

The γ-activity coming from ^{173}Lu is quite high. Some problems in the sagging of the photomultiplier gains were observed last two years in the ^{173}Lu experiments and also in ^{152}Eu experiment where a 3 x 10^{7} Bq high-energy γ-ray target was used. We have experienced that photomultiplier gains could be close to the nominal values with a 68 μg/cm^{2} ^{173}Lu target and 5 mm thick W liner (1.84×10^{7} γ/s).

We would thus like to keep the activity level seen by the DANCE array to around 10^{7} Bq or less. To this end we will use a 10 mm lead liner that surrounds the target to attenuate the low-energy γ-rays. The Pb liner would significant reduce the number of low-energy (up to ~200 keV) γ-rays, but there is little reduction for higher energy γ-rays (see tables 2-4 and Figure 5). To keep below our operating limit of 10^{7} Bq in case of 5 mm lead liner, we must reduce the target mass by approximately a factor of 3 (e.g., to 0.066 mg/cm^{2} target, similar than the previous target used in 2009).

Exploratory runs

As mentioned above, we will use a lead liner to reduce the activity level arising from low energy γ-rays. A thickness of 10 mm seems to be necessary but we may also need to try a 8 mm liner in order to keep the gamma activity below 10^{7} g/s.



Drawing 1: New lead liner (black), Al can (gray) and new RTH (center part) for the ^{173}Lu target

It will be necessary to test this lead liner in DANCE to check that our background due to scattered neutrons is not significantly increased. This lead liner was modeled by simulation using GEANT4.

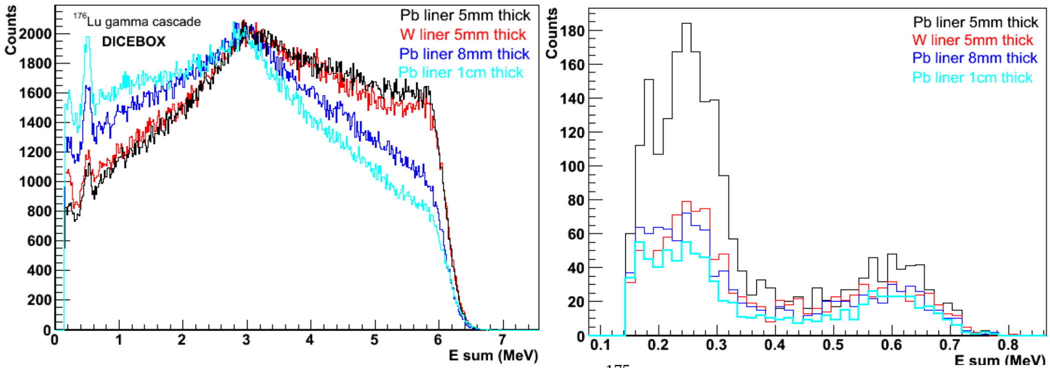
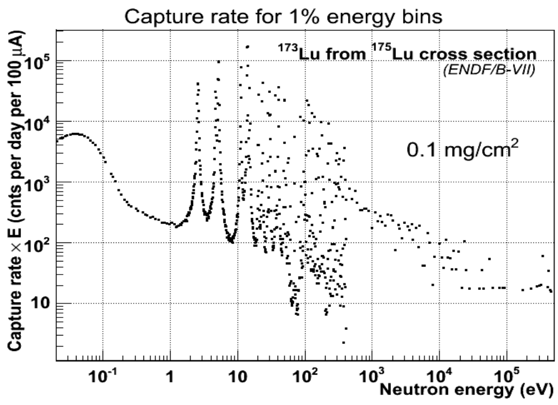


Figure 5: GEANT4 simulation results for E sum obtained from ^{175}Lu DICEBOX gamma-cascade (left) and from ^{173}Lu gamma decay (right). The effect of the lead liner is compared for various thicknesses and for the 5mm W liner we have used for the previous experiments.

To this end, we will design a new DANCE target holder that will fit inside the lead liner and the beam line. Also, as was done in the ^{152}Eu experiment we will need to change the operating conditions for the DANCE array (such as reducing the bias on the PMTs) and run a nonstandard data acquisition configuration as was done last year for ^{173}Lu test-experiment. For this, we request 2 days of beam time to test and optimize DANCE.

c. (n,γ) measurements

No data exist on ^{173}Lu. As the spin/parity of ^{173}Lu ground state is $7/2^+$ which is the same as ^{175}Lu, we assume that to first order the neutron capture cross sections will be similar. Therefore, the ^{173}Lu capture rate per day is calculated from the ^{175}Lu cross section and this estimate is shown in the figure 6a. Notice that as the working conditions of DANCE detector are affected by the high activity of the target and the effect of the lead liner, the resulting distorted response of DANCE is complicated to take into account in these calculations.



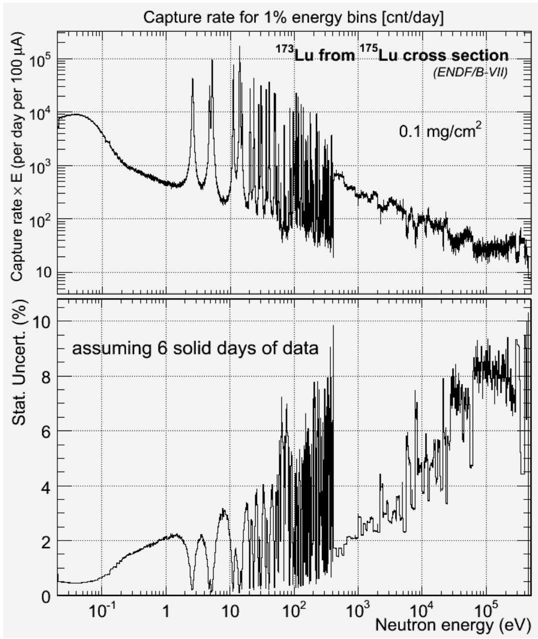


Figure 6a *The dotted line shows the count rate (counts per day) expected at the DANCE detector in 1% neutron-energy bins. This was obtained by folding the FP14 beam flux with the ENDF/B-VII cross sections for ^{175}Lu capture. Estimation for capture rate for 0.1 mg/cm^2 of ^{173}Lu.* ***Figure 5b:*** *The black line shows the count rate expected at the DANCE detector in 1% neutron-energy bins including a background due to the Ti backing. This was obtained by folding the FP14 beam flux with the ENDF/B-VII cross sections for ^{175}Lu capture and data from the blank target used for ^{175}Lu runs. Estimation for capture rate for 0.1 mg/cm^2 of ^{173}Lu. Statistical uncertainties are shown for ^{173}Lu.*

The statistical uncertainties are calculated assuming 6 solid days of data collection. In addition, we have included a background estimate taken from a blank target run that was collected last year. Figure 6b shows both the neutron capture rate and estimated statistical uncertainties for 1% energy bins including this background.

d. Total beam time request

Assuming that a new holder will be made for this experiment and taking into account the high activity level of the target, we need to have 2 days for feasibility tests with and without a Pb liner and to optimize the performance of DANCE under high count rate conditions. To obtain adequate statistics on the radioactive ^{173}Lu target itself, we request 8 days (7 days+1 day contingency). Six additional days are included to run blank and background measurements in addition to running Au as a calibration standard. A total request of 16 days is requested during the 2011 running period. This request includes 20% contingency due to beam deliver problems that can be expected from past experience.